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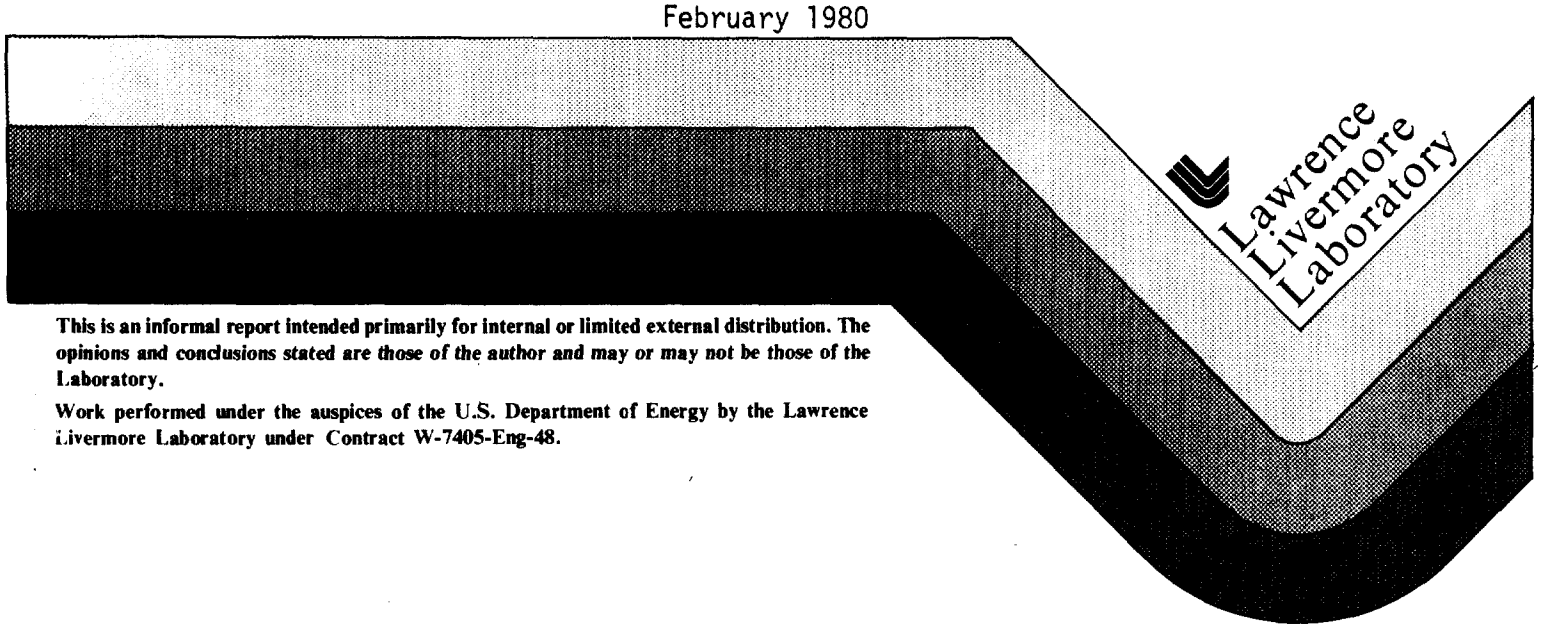

RESUSPENSION STUDIES AT BIKINI ATOLL

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ABSTRACT

The following experiments were conducted on Bikini Atoll to provide key parameters for an assessment of inhalation exposure from plutonium-contaminated dust aerosols: (1) a characterization of background (plutonium activity, dust, plutonium, sea spray, and organic aerosol concentrations), (2) a study of plutonium resuspension from a bare field, (3) a study of plutonium resuspension by traffic, and (4) a study of personal inhalation exposure. Studies similar to (1) and (2) have been previously performed at Enewetak Atoll.

Dust concentrations of 21 μg m^{-3} and sea spray of 34 μg m^{-3} were the background throughout Bikini Island except within 50 m of the windward beach. Background concentrations of $^{239+240}Pu$ were 60 aCi m^{-3} in the coconut grove and 264 aCi m^{-3} over rain-stabilized bare soil. The ratio of plutonium activity in aerosols (pCi g^{-1}) relative to the activity in underlying soil (pCi g^{-1}), defined as the enhancement factor, EF, was typically less than one apparently due to dilution by non-contaminated soil aerosols at a ratio of 1.8:1. Enhancement factors increased about 3.8 as a result of tilling on both Bikini and Enewetak. Plutonium resuspension flux was estimated at 0.49 pCi m^{-2} $year^{-1}$ over most of Bikini Island. Aerosol size distributions associated with mass and with plutonium activity were typically log-normal with median aerodynamic diameter 2.44 μm, which decreased to 2.0 μm above freshly tilled soil. The Pu concentration in aerosols collected over disturbed soil increased by a factor of 19.1 due to increased dust aerosol concentrations and increased enhancement factors. Vehicular traffic produced dust pulses typically of 10 s duration, 28 μg m^{-3} average concentration, and plutonium enhancement factor 2.5. Personal dosimetry showed that enhancement of dust by a worker stirring his own immediate environment was a factor of 2.64 when doing heavy work outdoors near the ground and 1.86 when doing light work in and around houses.

Pulmonary deposition of plutonium was calculated for various exposure conditions as determined by inhalation rate, aerosol dust concentration, plutonium activity in soil, plutonium enhancement factor, personal dosimeter enhancement factor and the pulmonary respirable-fraction. The pulmonary deposition ranged from 1476 aCi h^{-1} to 12 aCi h^{-1} with intermediate values for heavy outdoor work (139 aCi h^{-1}) and for light work in and around houses (78 aCi h^{-1}) plus a value of 1.58 aCi h^{-1} at roadside when passed by a typical vehicle once per hour.

INTRODUCTION

A study of inhalation exposure from plutonium-contaminated soil was conducted to provide the parameters for a rigorous assessment of the inhalation dose to man. The study was needed because in previous dose assessments only a minimum amount of information was available about total mass-loading of suspended aerosol (μg/m^3),

aerosol size, and total radioactivity per unit mass. Although necessary, these parameters are insufficient to describe the fraction of suspended radioactivity within the respirable size range and how either the concentration of radioactivity or the respirable fraction vary with surface conditions and local resuspending (dust-lifting) mechanisms.

Our investigations recently conducted on Bikini and Enewetak Atolls in the Marshall Islands provided new data which have implications not only for the local dose assessment concerning rehabitation of those sites, but which are important for understanding low-level inhalation exposure to toxic radionuclides in general.

BACKGROUND

This study conducted on Bikini Island in May 1978 provided a more complete set of data, following our preliminary studies on Engebi Island of Enewetak Atoll in February 1977. The Bikini Island study utilized extensive soil sampling and in situ gamma spectroscopy to determine isotope concentrations in soil and vegetation. Also various air sampling devices were used to determine particle size distribution and mass loading, and micrometeorological techniques were used to determine aerosol fluxes. Subsequent wet chemistry analysis provided radionuclide and elemental concentrations in collected aerosols. Four simultaneous experiments were conducted: (1) a characterization of the normal (background) suspended aerosols and the contributions from sea spray off the windward beach leeward across the island, (2) a study of resuspension of radionuclides from a field purposely laid bare by bulldozers as a worst-case condition, (3) a study of resuspension of radioactive particles by vehicular and foot traffic, and (4) a study of personal inhalation exposure using small dosimeters carried by volunteers during their daily routines. Less complete studies similar to (1) and (2) had been performed previously on Engebi Island at Enewetak Atoll and background studies similar to (1) were performed later on Eneu Island at Bikini Atoll.

METHODS

Soil and vegetation samples were collected for analysis of radionuclide concentrations. ^{238}Pu, $^{239+240}Pu$, and ^{241}Am concentrations were determined by isotope dilution and alpha spectrometry and ^{90}Sr concentrations by yttrium-90 separation and beta counting. These analyses were performed by LFE Corporation. Gamma spectroscopy using Ge(Li) diode detector was used to determine ^{137}Cs concentrations. Also, because the ratios $(^{241}Am)/(^{239+240}Pu)$ and $(^{238}Pu)/(^{239+240}Pu)$ are constant on Bikini, it was possible to estimate plutonium soil concentrations by measuring ^{241}Am soil concentrations using a gamma spectroscopy system consisting of a planar, high-purity germanium diode which was cryogenically cooled to achieve a minimum detectability for ^{241}Am less than 1.0 pCi g^{-1} (1). The detector was mounted facing downward on a tripod so that the volume of soil integrated was contained in a circle of probable detection of nominally 3 m radius and 5 cm depth. Because the nuclear events causing the original contamination of Bikini Island were far removed, the fallout was relatively evenly dispersed across the Island.

Impingers were used to collect soluble sea spray aerosols in a 250 ml distilled water trap similar to the method of Hsu and Whelan (2). Air flow rates were 0.36 m^3 h^{-1} (6 l min^{-1}) through the water trap and measured amounts of water were added each day to replace evaporated water (nominally 40 cm^3 day^{-1}). Impingers

were set at four tower locations along a 60 m wide clearing from the windward beach inland, spaced at 3, 26, 52, and 102 m from the high-tide waterline and at 1 m and 4 m above the ground. Elemental analysis on the remaining water was obtained by inductively-coupled plasma-optical emission spectroscopy for Na, K, Mg, Ca, and Zn and by a standard autoanalyzer (Technicon) for Cl.

The major particle collection system was an array of 14 standard-filter, high-volume air samplers (HV) and two cascade impactors both using Gelman type AE glass fiber filters. Flow rates for HV were monitored at a pressure tap on the fan; discharge was nominally 100 $m^3\ h^{-1}$ (60 cfm). The lapsed time of filter operation was recorded for each HV and cascade impactor by counting pulses from a crystal-controlled clock activated by a pressure sensitive switch. Cascade impactors (CIP) were the 5-stage, jet-plate type (Model 65-000, Anderson Air Samplers 2000 Inc.). Three HV with the air inlets at 1.1 m above ground were located on a line from the windward beach inland at 5, 70, and 158 m from the high-tide waterline; the latter two were beneath a coconut grove canopy. One HV was located to the lee of a road at the traffic study site, and ten HV and two cascade impactors were placed in a square array nominally 10 m apart in the middle of a field (one hectare area) cleared by bulldozing. Special chemistry methods (LFE Corporation) were employed on the filters to determine the concentration of the stable elements Na, K, Ca, Mg, S and Cl, and the radioactive isotopes ^{238}Pu, $^{239+240}Pu$, ^{90}Sr, ^{137}Cs, and ^{241}Am. Filter blanks were used to correct the stable elements. See map, Figure 1, for locations of instruments.

In addition to the HV, three non-standard ultra-high volume air samplers (UHV) were used having air inlets at 1.5 m height. Flow-rates, nominally 2550 $m^3\ h^{-1}$ (1500 cfm), were monitored both by a pitot tube pressure tap on the fan discharge and by a modified anemometer transducer measuring total discharge in the fan outlet which is ducted 4 m downwind before discharge. Filters were 1 m^2 area of special fiber-type (Microsorban N-98, Aerosol Filter Grade S, Delbag Luftfilter). One UHV was placed in the coconut grove 370 m from the windward high-tide waterline, and two UHV were operated in the cleared field at the downwind edge of the HV array. The UHV provided the advantage of detection of suspended radioactive aerosols at extremely low levels (e.g., worldwide background) in a matter of a few hours run time. (Locations of UHV are shown on Figure 1).

Personal dosimeters (Model S, Monitaire, Mine Safety Appliances Co.) were used to determine exposure rates of individual persons to suspended particles. The personal dosimeters (PD) are small, belt-mounted pumps with a hose connection to a cyclone particle discriminator and filter holder suspended by a chain worn around the neck. Flow rates are 0.10 $m^3\ h^{-1}$ (0.06 cfm) and filters were porous-type membrane filters (37 mm diameter, 1 μm pore diameter, Nuclepore Corporation). It was found that blank membrane filters inexplicably gained weight with time but that standard deviations within 10% of mass could be achieved where filter blanks from the same lot were monitored for weight gains over the time period of the experiment. The membrane filters were used as a substrate for a scanning electron microscope (SEM) study of particle characteristics. The SEM operated by the LLL Particle Characterization Facility has a large chamber for specimens (90 mm) and has a resolution of 0.015 μm. In the SEM mount, microprobe chemistry of individual aerosol particles on the membrane filters was accomplished by X-ray fluorescence with a resolution of 160 eV, which provided quantitation of particles containing elements with atomic numbers equal or greater than sodium.

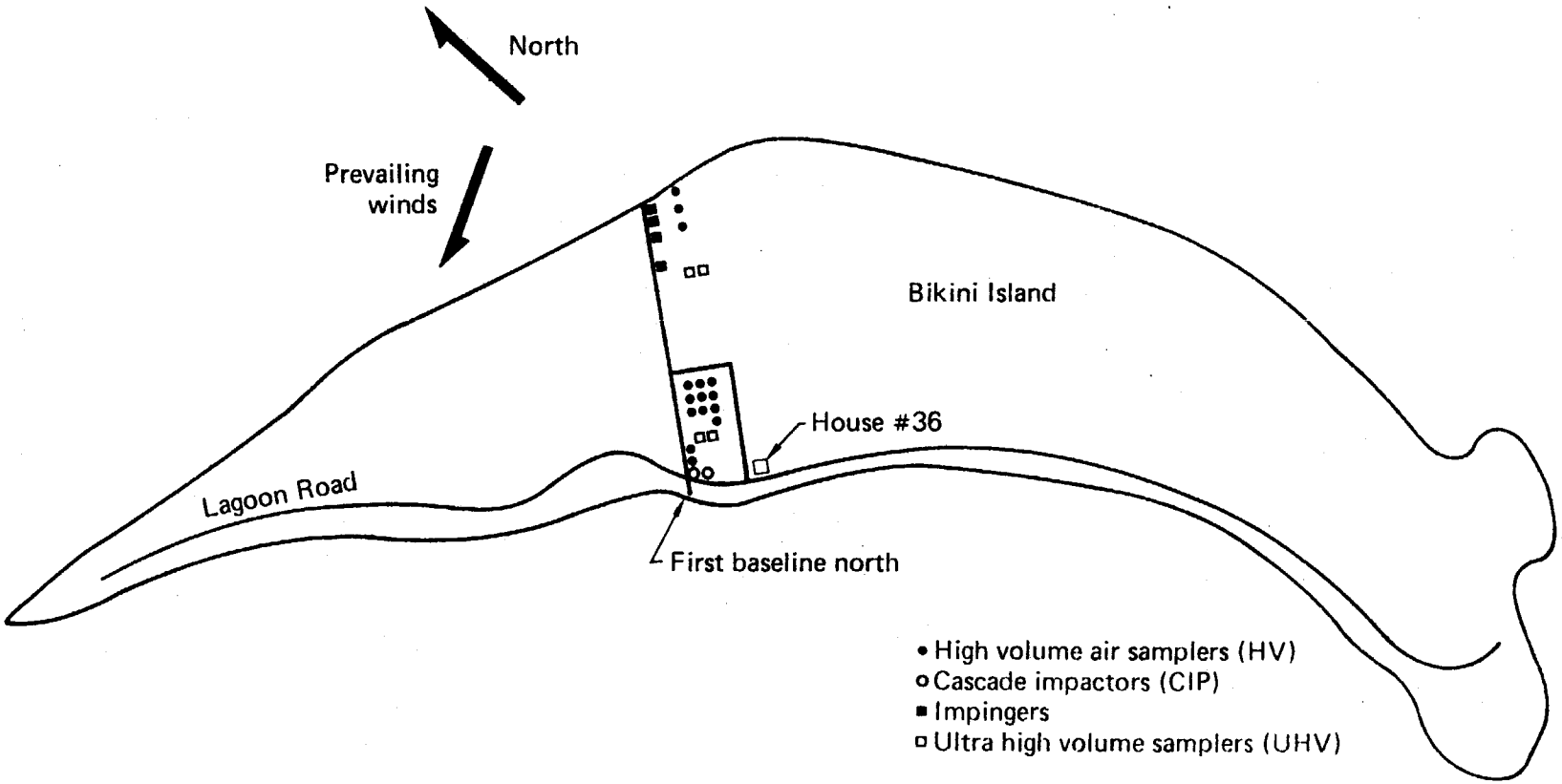

Figure 1. Plan View of Instrument Locations on Bikini Island, 1978.

Light scattering instruments capable of sizing and counting particles in the diameter range 0.3 μm to 10 μm were used in two modes. In the first mode, a particle analyzer (Model CI-201, Climet Instruments Co.) was operated so that in situ particle-size number-densities were determined. Particles in air entering the view volume at a rate of 0.43 m^3 h^{-1} (0.25 cfm) are counted individually by the resulting pulses which are a linear function of particle diameter and classified by means of a conventional 200-channel pulse-height analyzer at a resolution of about 20 channels μm^{-1} (latex sphere calibration). Particle sizes were measured in this mode at the 1.0 m height in the cleared field. Particle counts were integrated over 120-second periods at frequent intervals during the experiment. In the second mode, light scattering instruments are used as continuous monitors of total particle concentration. Two instruments were used in this mode: one, similar to the above but without size discrimination (Model CI-208A, Climet Instruments Co.), was also placed at the 1.0 m height in the cleared field. The second instrument used as a mass concentration monitor was an integrating nephelometer which measures the bulk scattering coefficient. In the one micro-meter size range, the nephelometer samples ambient air at a flow rate of 17 m^3 h^{-1} (10 cfm) and has a fast response (about 1 sec) with a continuous DC analog output that is linearally related to mass concentration using the manufacturer's calibration (Model 1560, Meteorology Research Inc.). On Bikini, the nephelometer was used as a dust monitor for foot and vehicular traffic.

Two, portable, wind systems were utilized. For monitoring wind speed and direction in the open field at the 4.5 m height, a wind speed transmitter with reed switch contact closure each revolution and a wind direction transmitter with 540° dual potentiometer were operated through a battery-powered translator with 30-second filter, and recorded on a dual channel, strip chart recorder with an 8-day spring-driven chart. Wind speed vertical profiles for aerosol flux estimates were determined using a portable, sensitive, cup-anemometer system (modified, from C.W. Thornthwaite Associates) operated at five height levels up to 4 m for selected 15-minute periods (stall speed 0.20 m s^{-1}, distance constant 1.1 m).

RESULTS AND DISCUSSION

Characterization of Background Aerosols

The contribution of sea spray to aerosol mass loading was investigated thoroughly because it is a diluent of the resuspended mass fraction from the soil which is the host to plutonium in the atmosphere. The stable element analyses showed that the ratios Cl/Na and Na/Mg were conservative and had values predicted by the composition of seawater (1.8 and 8.3, respectively) (3). Other elements, in particular, Ca, were expected to be a tracer for calcareous soil from the parent coral material. The ratio Na/K was somewhat more variable and had a mean value of 11 which is intermediate between those values predicted for sea spray (5.7) and for seawater (27) (3, 4). Hence, the concentration of sea spray in the air was calculated by 3.25 x Na concentration and 27 x Mg concentration, based on the ionic composition of seawater (4). These two values were averaged to minimize random error of determination. The impingers were determined to be 77% efficient for collection of the sea spray by comparison with HV; after this correction and averaging the samples from 1 m and 4 m heights, it was found that the concentration of sea spray was constant over the island to within 52 m from the windward, high-tide waterline (Table 1). The leeward decrease was verified by HV measurements

TABLE 1. Variation of Na and Mg aerosols and sea spray with distance from the waterline (impingers, effectively at a 2 m height).

Distance (m)	Na ($\mu g\ m^{-3}$)	Mg ($\mu g\ m^{-3}$)	Calculated Sea Spray ($\mu g\ m^{-3}$)
3	56.2	6.17	175
26	13.3	1.45	41
52	10.5	1.21	35
102	10.4	1.30	34
Background (HV)	10.5	1.26	34

as well, but it occurred in a surprisingly short range compared to sea spray aerosols measured by other investigators (2,5,6). The rapid drop-off of sea spray from the shoreline is thought to be due to the presence of a massive vegetative barrier along the shore and we expect that the horizontal flux is already reduced at the shore because the sea spray is mostly generated at the surf line on the coral reef nearly a kilometer upwind from the beach. The HV measurements show that the background sea spray aerosol calculated from Na and Mg concentrations was remarkably uniform throughout the remainder of the island ($\bar{x}$ = 34 $\mu g\ m^{-3}$, S = 8.7, n = 27). The HV results are summarized in Table 2.

Background is here defined as the aerosol concentrations at the 1.1 m height over surfaces which are relatively stabilized and under normal wind conditions. After a week, even the bare soil tended to reach the same average level of dust aerosol concentrations (21 $\mu g\ m^{-3}$) as the coconut grove (Table 2). An analysis of the personal dosimeter data (discussed later) showed that about 10% of the background dust aerosol was organic. $^{239+240}Pu$ concentration (aCi/m^3) was a factor of 258/60 = 4.3 greater over bare soil than in the coconut grove. (Soil activity was 15.3 and 8.01 pCi/g respectively, which is not significantly different within the normal variation encountered.)

If we examine the vertical fluxes of plutonium ($aCi\ m^{-2}\ s^{-1}$), the ratio of fluxes from the two sites will be proportional to the ratio of their wind friction velocities, u_*, where:

$$u_* = C_D\ U_1 \qquad [1]$$

and C_D is a drag coefficient equal to 0.106 in the coconut grove and 0.077 in the bare field as determined by our wind profile measurements and U_1 is the wind speed at the 1 m height, which was 4.1 x greater for the bare field than the coconut grove. By Equation 1, the ratio of friction velocities is 3 x greater in the bare field than in the coconut grove. The ratio of their plutonium fluxes is also proportional to the ratio of their concentrations; hence, the plutonium flux is a factor of 4.3 x 3 = 12.9 greater in the stabilized bare field than in

TABLE 2

SUMMARY OF MASS AND PLUTONIUM AEROSOL CONCENTRATIONS ON BIKINI ISLAND

DATE 1978	DISTANCE TO WINDWARD SHORE (m)	GROUND COVER	SEA SPRAY ($\mu g\ m^{-3}$)	DUST AEROSOL ($\mu g\ m^{-3}$)	PLUTONIUM CONCENTRATION ($aCi\ m^{-3}$)*	TYPE (number) OF INSTRUMENTS	WIND ** SPEED ($m\ s^{-1}$)	WIND ** DIRECTION (deg)
5/6-8	780	Bare Soil	--	167	7295	CIP (2)	4.7	53
5/9-16	780	Bare Soil	--	9	246	CIP (2)	4.6	43
5/6-8	600-700	Bare Soil	34	136	6466	HV (3)	4.7	53
5/10-11	600-700	Bare Soil	34	23	338	HV (10)	4.1	52
5/12-16	600-700	Bare Soil	35	18	189	HV (10)	4.6	33
5/8-16	70-160	Coconut Trees	34	21	65	HV (2)	4.6	45
5/8-16	820	Road	33	41	421	HV (1)	4.6	45
5/8-16	5	Shrubs	40	8	29	HV (1)	4.6	45
5/10-11	370	Coconut Trees	--	--	51	UHV (1)	4.1	52
5/12-16	760	Bare Soil	--	--	212	UHV (2)	4.6	33
Background ***	>50	Bare Soil	34	21	258	CIP(2),HV(20),UHV(2)		
Background ***	>50	Coconut Trees	34	21	60	HV(2),UHV(1)		

* aCi (attocuries) = 10^{-18} Curies

** Wind measurements recorded for the 4.5 m height at a station in the open field.

*** Averaged over the surfaces that are stabilized.

the coconut grove. In our previous work, we calculated plutonium aerosol flux with the equation,

$$F = -pk\, u_* \chi_1 \qquad [2]$$

where p is the exponent of a presumed power-law distribution of Pu with height (negative sign indicating decreasing concentration with height), k is Karman's constant equal to 0.4, and χ_1 is the plutonium concentration in the height range from 0.5 to 2.0 m (7). The exponent p is the slope of the Pu concentration versus height on a log-log scale. The impinger measurements at the 1 m and 4 m heights along a 60 m wide clearing parallel to the mean wind direction showed a p-value of 0.55 for calcium on Bikini Island which we presume is the major host of terrestrial Pu contamination. Previous work indicates p values between 0.25 and 0.35 for dust aerosols in Western U.S. (7). Using the local p-value of 0.55 and measured values of u_* and the background plutonium concentration, χ_1, (60 aCi m^{-3}) as typical for the coconut grove we obtain a plutonium resuspension flux of 1.54 aCi m^{-2} s^{-1} (0.49 pCi m^{-2} $year^{-1}$) which compares to 19.9 aCi m^{-2} s^{-1} (6.3 pCi m^{-2} $year^{-1}$) from the stabilized bare field. This discussion of flux cannot explain why the dust aerosol concentration is the same at both sites, but presumably the mixture of calcareous aerosols, organic matter, and other components of dust are quite different for each site.

Resuspension of Radioactive Particles from a Bare Field on Bikini Island

On May 6, 1978, a field was chosen for convenience (adjacent to House No. 36) and bulldozed bare of vegetation without stripping the soil. At the windward end of the 100 m x 200 m field, the array of instruments (10 HV, 2 CIP, and 2 UHV) were set up in a regular grid covering about one hectare. The upwind fetch to the nearest instrument was 60 m and lateral borders were 30 m wide. During May 6-8, three HV and two cascade impactors (CIP) were run during the highest resuspension (disturbed) phase immediately after bulldozing, followed by extensive runs with all instruments during the stable phase, May 9-16. Wind speeds and direction remained relatively constant (Table 2). Plutonium aerosol concentration (aCi m^{-3}) was increased in the period May 6-8 over the period May 9-16, by a factor of 25 to 30 as shown by the HV and CIP data of Table 2. Because the disturbed surface was stabilized by light rain at the end of the run on May 8, the cascade impactor data showed significant differences in the plutonium-activity size distribution as shown on Figure 2 (ordinate $d\chi/d \ln D$ in units pCi g^{-1} of dust aerosol). The plutonium activity curves of Figure 1 are calculated log-normal distributions with the median aerodynamic diameters (MAD) and geometric standard deviations (GSD) obtained by fitting cascade impactor data (Table 3). The aerosol size distributions for plutonium activity determined by CIP and the total mass loading (sea spray plus dust) determined by optical particle analyzer were satisfactorily approximated by a log normal distribution with the given GSD values in Table 3. All other MAD values of Table 3 were determined by cascade impactor, but more data would be required to determine if a log normal distribution exists for the other aerosol definitions.

Two typical cases of number density ($dN/d \ln D$) and volume density distributions ($dV/d \ln D$) determined by the optical particle analyzer over the stabilized bare soil surface on May 9 and May 11 are shown in Figure 3. It should be noted that the optical particle analyzer sees all liquid and solid aerosols including

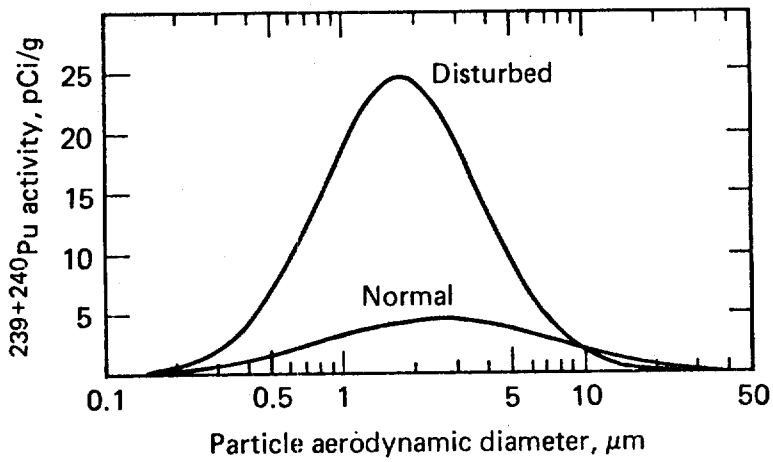

Figure 2. Plutonium activity (pCi g^{-1}) versus particle aerodynamic diameter D (μm) at the 1.1 m height over bare soil on Bikini Island (wind speed 4.5 m s^{-1}, surface contamination 15.3 pCi g^{-1}).

TABLE 3. Aerosol size characteristics on Bikini Island determined by cascade impactors and the optical particle analyzer at a height of 1.1 m.

Median Aerodynamic Diameters (μm)	Disturbed Bare Soil	Stabilized Bare Soil
Pu Activity (pCi g^{-1})	1.73	2.46
Pu Concentration (aCi m^{-3})	2.05	2.43
Mass Loading (μg m^{-3})	2.03	2.46
Mass Loading-Optical (μg m^{-3})	--	2.40 (.11)*
Sea Spray - Mg (μg m^{-3})	--	2.59
Geometric Standard Deviation		
Pu Activity (pCi g^{-1})	2.16	3.09
Mass Loading - Optical (μg m^{-3})	--	2.82 (.25)*

* Optical particle analyzer data with standard deviations in parentheses.

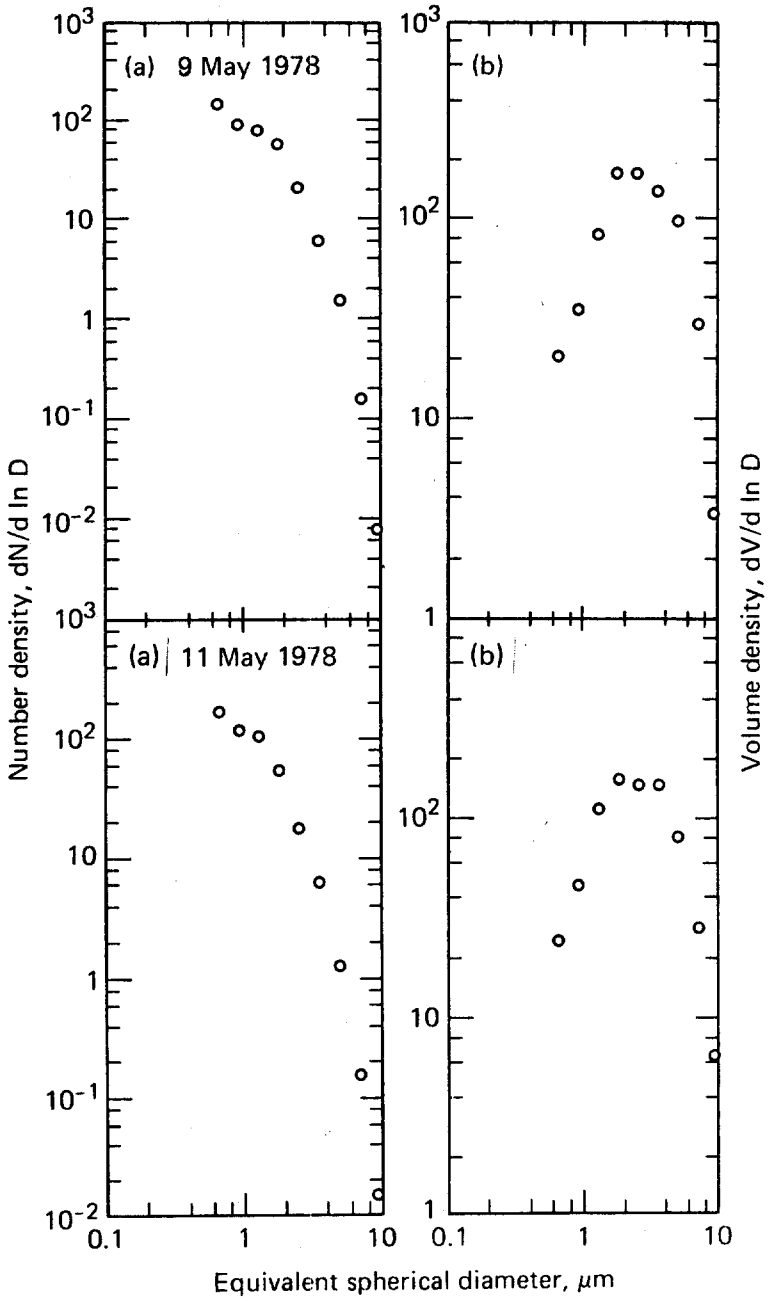

Figure 3. Typical particle size distributions over the stabilized bare soil surface. Number density (a) has units of particles cm^{-3}; volume density (b) has units μm^3 cm^{-3}.

both dust and sea spray so that the total mass obtained by integrating the volume distribution and multiplying by a density factor would not be expected to agree with the dry, residual mass loading measured by HV and CIP.

The relatively good agreement obtained between the different measurement systems indicated in Tables 1, 2, and 3 gives us the confidence to draw conclusions about the significance of resuspension for enhanced inhalation exposure in this worst case example. If the total plutonium activity (pCi g^{-1}) is obtained by integration of the curves of Figure 1, we find that there is a significant change in the aerosol plutonium activity relative to the plutonium activity of the surface soil. Let us define an enhancement factor (EF) as follows:

$$EF = \frac{\text{total aerosol activity (pCi } g^{-1})}{\text{soil activity (pCi } g^{-1})}$$

Upon investigation of the enhancement factors, we find that the values apparently were less than one under normal conditions (Table 4) probably because of selective resuspension of non-radioactive particles which served to dilute the plutonium activity. There is no evidence how this occurred. Martell surmises ratio values should be greater than one, but gives no evidence (8). The ratio of organic particles to calcareous soil particles remained about constant (10%) as determined by X-ray fluorescence on the PD filters exposed at this site during the same period. We know from previous studies that one component of organic matter, plant leaves, had a ratio of 10^{-3} plutonium concentration relative to soil and could serve to dilute the inorganic aerosol.)

TABLE 4. Enhancement factors for plutonium activity of aerosols on Bikini Island (HV data).

Site	Date	Soil Activity (pCi g^{-1})	Aerosol Activity (pCi g^{-1})	Enhancement Factor
Disturbed bare soil	5/6-8	15.3	47.5	3.10
Stabilized bare soil	5/10-11	15.3	14.7	0.96
Stabilized bare soil	5/12-16	15.3	10.5	0.69
Coconut grove	5/8-16	8.01	3.29	0.41
Road with traffic	5/8-16	4.10	10.3	2.5

Under dusty conditions, EF values exceed one such as in the cases of the disturbed bare soil (3.1) and the road with traffic (2.5). So there are two different factors producing increased plutonium aerosol concentrations (aCi m^{-3}) during unusual resuspension. The aerosol dust concentration increases, but also the plutonium activity increases. For example, averaged over 10 HV instruments,

the ratio of plutonium concentration over bare soil on May 6-8 compared to May 10-11 is caused by a 5.91 x increase in dust aerosol concentrations (Table 2) and 3.23 x increase in enhancement factor (Table 4) for a combined effect on aerosol plutonium concentration (6466 aCi m^{-3}/338 aCi m^{-3}) of 19.1.

Resuspension of Radioactive Particles by Vehicular and Foot Traffic

The integrating nephelometer was installed with intake at 1.2 m height and 2 m leeward from the position of average tire tracks on a frequently-traveled, one-lane dirt road on Bikini Island. Even though the traditional vehicular traffic of light trucks at low speeds was increased in frequency by our experimental activity, we were interested in characterizing the resuspension of plutonium and inhalation exposure per vehicle pass. The nephelometer provided details on magnitude, duration, and frequency of dust concentrations, while plutonium and dust aerosol concentrations (Table 2), and plutonium activity and enhancement factors (Table 4) were obtained by a co-located HV.

Dust concentrations above background rose in a pulse exceeding 10 s duration where the peak was obtained in a period about 4.5 s after the passage of the vehicle (Figure 4). This characteristic time to arrival of the peak, regardless of concentration, was determined by X/σ_u where the travel distance X is 2 m and the RMS turbulent velocity σ_u is about one-tenth the local wind speed of 4.5 m s^{-1}. Hence the dust pulse was traveling by diffusion and not characterized by translation in the wake of the passing vehicle. The dust pulse example of Figure 4 represents an extreme case (more than 90% of occurrences had lower concentrations), but demonstrates the characteristic peak to mean ratio of 3.6 and the slow return to background on the tail of the pulse. The amplitude and frequency of dust pulses due to motor vehicle, bicycle, and foot traffic were recorded during May 11-15. The sixty-eight cases of motor vehicle passes observed showed an approximate log-normal frequency distribution with median peak concentration (above background) of 100 μg m^{-3} and geometric standard deviation of 3.4, Figure 5. Bicycle traffic could not be distinguished from foot traffic. In the seven observed cases of foot traffic, we found an approximate median peak concentration above background of 26 μg m^{-3}.

It should be emphasized that the log-normal concentration implies a fairly high chance (5%) of an exposure to a vehicular-induced peak concentration of 760 μg m^{-3} having a mean concentration 760/3.6 = 211 μg m^{-3} for about 10 seconds. The plutonium enhancement factor was estimated at 2.5 in this study (Table 4).

Personal Inhalation Exposure and Dosimetry

Until now, the discussion has centered on the (combined) isotope $^{239+240}Pu$, since in fact this is the most important component of inhalation exposure. Extensive soil sampling on Bikini Island has established that a relatively homogeneous mixture of isotopes exists in the soil (Table 5). In the aerosols, some of the isotopes become significantly enhanced (^{238}Pu and ^{137}Cs) but they remain of lesser inhalation-hazard. The in situ gamma (ISG) spectroscopy system which measured ^{241}Am in the soil at Bikini was highly correlated to that measured in surface soil samples by special chemistry methods (r^2 = 0.910), which gives confidence in both methods. However, data from the ISG system was consistently lower than the soil sampling method by a factor of 0.7 because it integrated a

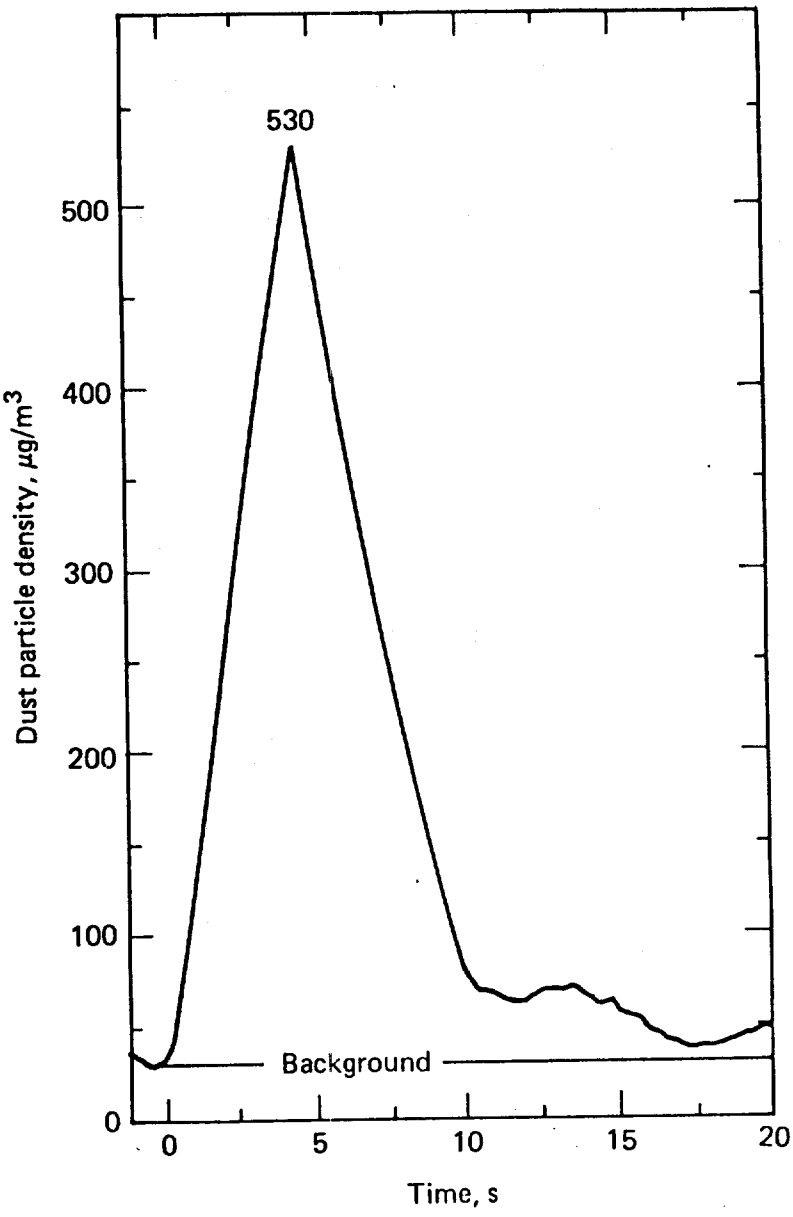

Figure 4. An example of a dust concentration pulse on the downwind side of a one-lane dirt road following passage of a light motor vehicle, 11:05 a.m., May 15, 1978 (wind speed 4.5 m s^{-1}).

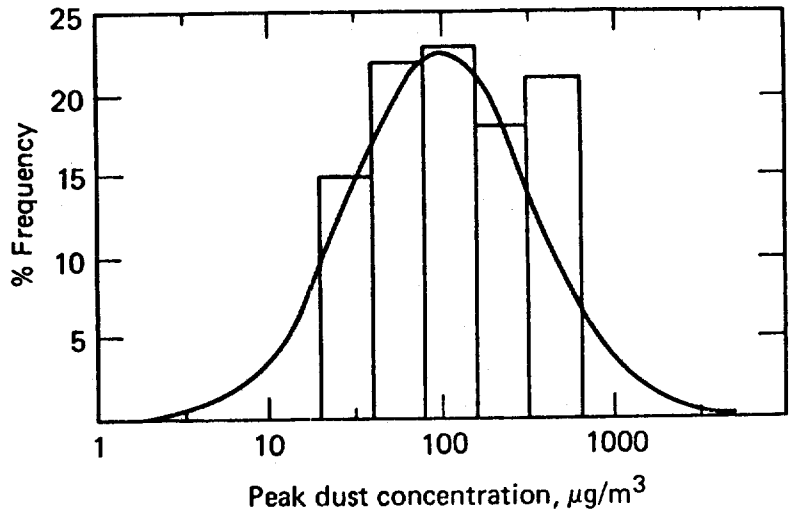

Figure 5. Frequency of peak dust concentrations on the downwind side of a one-lane dirt road following passage of light motor vehicle.

TABLE 5. Radioactive isotope ratios in soil and aerosols at a stabilized bare field on Bikini Island

	$\frac{^{238}Pu}{^{239+240}Pu}$	$\frac{^{241}Am}{^{239+240}Pu}$	$\frac{^{137}Cs}{^{239+240}Pu}$	$\frac{^{90}Sr}{^{239+240}Pu}$	$^{239+240}Pu$ (pCi g^{-1})
Soil	0.0013	0.556	9.80	9.15	15.3
FSD*	0.56	0.55	0.48	0.52	0.57
Aerosols (HV)	0.050	0.439	61.4	12.8	12.6
FSD*	0.64	0.34	0.32	0.51	0.35

*Fractional Standard Deviations ($s/\bar{x}$)

view volume about 5 cm depth and the exponential decrease of isotope concentration with depth gives lower mean values. After correction by a factor 1.44, the ISG method was the primary method for mapping ^{241}Am as a tracer for the source of suspended plutonium isotopes. The horizontal variations in soil isotope concentrations (ISG data) were small enough so that one could justify mean values as local regional values for soil. For example, on the one-hectare, bare field, it was determined that plutonium in surface soil had the mean value 15.3 pCi g^{-1} with an observed range of 2.3 to 28, and a fractional standard deviation ($s/\bar{x}$) of 0.57. There was no apparent pattern to the soil concentrations and they exhibit approximately normal, random variation perhaps due to previous tilling and blading locally. (The data did not fit a log-normal distribution any better.)

In the context of this surface soil contamination, personal dosimeters (PD) provided information about inhalation exposure of individuals relative to the reference HV monitors. It was found that the fraction of dust in the total aerosol collected by the PD was greatest for workers exposed during heavy tilling but was also high for workers exposed in and around houses (Table 6) partly because of a lowered fraction of sea spray in both cases. In this and prior studies, we found that the ratio of PD Dust/HV Dust has a value of approximately 0.5 where both PD and HV are sampling the same aerosol cloud of this size particles (2.5 μm MAD) because of the cyclone particle-discriminator on the PD. Therefore, the enhancement of inhalation exposure by a worker's own actions where the PD and HV are <u>not</u> sampling the same cloud can be estimated by a personal dosimeter enhancement (PDE):

$$PDE \simeq 2x \text{ (PD Dust/HV Dust)} \quad . \qquad [4]$$

Values so computed show significant enhancement (PDE) of inhalation exposure (2.64) during heavy work outdoors by persons sitting or kneeling while digging or using tools on the ground (Table 6). The second highest enhancement (1.86) came from persons with duties in and around the houses. Other work, including

TABLE 6

Comparative Analysis of Personal Dosimeters (PD) Worn by Volunteers in Various Work Assignments

Activity	Outdoors Over Disturbed Bare Field	Outdoors Over Stabilized and Vegetated Surfaces		Inside and Outside Around Houses
	During Tilling	Light Work	Heavy Work	Light Work
Number of Volunteers	2	4	3	3
PD Dust Fraction *	94%	56%	56%	89%
PD Dust ($\mu g\ m^{-3}$)	62	12	28	20
PD Dust/HV Dust	.46	.55	1.32	.93
PD Enhancement **	.92	1.10	2.64	1.86

* Personal Dosimeter dust is corrected for sea spray but contains about 10% organic matter, both estimated by x-ray fluorescence.

** PD Enhancement = 2x (PD Dust/HV Dust).

heavy tilling, produced inhalation exposures satisfactorily monitored by HV and thus, their PDE values were close to unity. The main limitation of the PD data and the derived enhancement values (PDE) is that no information is obtained about the plutonium enhancement factors expressed by Equation [3]. It should be recalled that plutonium enhancement factors (EF) of the same magnitude as these PDE were detected by HV (Table 4).

Pulmonary deposition is the penetration and retention of respirable size particles into the deep, alveolar regions of the lung and constitutes the major vector of inhalation dose. The efficiency of pulmonary deposition varies with particle size and is conventionally estimated by the ICRP Task Group on Lung Dynamics' deposition model (9). For example, the observed change in size distribution of plutonium activity from the stabilized soil case to the disturbed soil case (Fig. 1), increased the calculated pulmonary respirable-fraction (RESP) from 19% to 24%. Pulmonary deposition (attocuries per hour) using an inhalation rate (IN. RATE) and previously defined terms may be estimated as follows:

$$\underset{(aCi\ h^{-1})}{DEPOSITION} = \underset{(m^3\ h^{-1})}{IN.\ RATE} \times \underset{(\mu g\ m^{-3})}{HV\ DUST} \times \underset{(aCi\ \mu g^{-1})}{SOIL\ ACTIVITY} \times EF \times PDE \times RESP \quad [5]$$

Using Equation [5], data from Tables 2, 4, and 6, and the best estimates for the enhancement factors, inhalation rate, and respirable fraction extrapolated from our measurements, we calculated pulmonary deposition of $^{239+240}Pu$ for four cases on Bikini (Table 7). Under the worst case condition (during tilling in a disturbed bare field), the pulmonary deposition was 1476 aCi h^{-1}, and in the best case (light work in a coconut grove), the pulmonary deposition was 12 aCi h^{-1}. Intermediate values were 139 aCi h^{-1} for heavy work in a bare field, and 78 aCi h^{-1} for light work in around houses. (In the latter case, we had to use an enhancement factor measured in the nearby field rather than in and around the houses.)

Walking along the road with one vehicular passage per hour produced an estimated 50% chance of additional pulmonary deposition of 1.58 aCi h^{-1} (above background) but the soil plutonium activity on the road was notably lower (4.1 pCi g^{-1}) compared to the field (15.3 pCi g^{-1}); see Table 7.

To put these estimated pulmonary deposition values in perspective, we have estimated the lung and bone doses from inhalation of $^{239+240}Pu$ and ^{241}Am using the ICRP lung model (8). The scenario we adopted is arbitrary; we assume that a person is in high activity conditions (1500 aCi/h pulmonary deposition) for 5 hours per day and in a situation averaging 80 aCi/h for the other 19 hours. The daily average is therefore 376 aCi/h pulmonary deposition. If it is assumed that people are exposed to this level everyday throughout their life, then the maximum bone dose rate is 5.5 mrem/y and the maximum lung dose rate is 2.4 mrem/y; the 30 year integral doses are 36 mrem and 70 mrem for bone and lung respectively. These doses are well below the Federal Guidelines for bone and lung of 500 mrem/y and 5 rem in 30 years. It is also quite possible that the selected scenario of 5 hours at the high activity deposition rate (1500 aCi/hr) is on the average a very high estimate of the annual time spent under such conditions.

TABLE 7

Pulmonary Deposition of Plutonium ($^{239+240}Pu$) for Worst Case and Best Case Conditions on Bikini.

Condition	Inhalation Rate ($m^3\ h^{-1}$)	Dust Aerosol ($\mu g\ m^{-3}$)	Soil Pu Activity ($aCi\ \mu g^{-1}$)	Enhancement Factor (EF)	Personal Enhancement (PDE)	Respirable Fraction (RESP)	Pulmonary Deposition ($aCi\ h^{-1}$)
Bare Field, During tilling	1.04	136	15.3	3.10	0.92	0.24	1476
Stabilized Field, Heavy work	1.04	21	15.3	0.83	2.64	0.19	139
In and Around Houses, Light work	0.83	21	15.3	0.83	1.86	0.19	78
Coconut Grove, Light work	0.83	21	8.0	0.41	1.10	0.19	12
At Roadside, One Vehicle/Hr *	0.023	28	4.1	2.50	(1.0)	0.24	1.58 + BG

* Exposure to one, ten-second, median, vehicular dust-pulse, not including background (BG).

SUMMARY AND CONCLUSIONS

Mass loading (all aerosols) on a HV filter was 55 μg m^{-3} on Bikini Island over stabilized and vegetated surfaces (e.g., in a bare field following rain and in a coconut grove). This compares to 56 μg m^{-3} measured at a vegetated site on Engebi Island of Enewetak Atoll in February 1977, and a 42 μg m^{-3} weekly average for 10 weeks in a coconut grove on Eneu Island of Bikini Atoll May-August, 1978. (Wind speeds were comparable, 4-5 m s^{-1}, in all cases.) The more detailed studies at Bikini revealed that 34 μg m^{-3} of the mass loading was salt from sea spray, and that this sea spray contribution remained constant across Bikini Island beyond 20-50 m from the windward beach.

The "background" concentrations of aerosol plutonium on Bikini are comparable to those on Engebi Island, Enewetak, when one considers the surface soil plutonium activity (Table 8). And, by assuming that Engebi Island had the same aerosol sea spray background (34 μg m^{-3}) as Bikini (which has not been verified by actual measurement) we found that the enhancement factors agree reasonably well. The normal enhancement factor is 0.56, if one assumes that values less than one (Table 8) represent normal variations about the mean of 0.56. Apparently, the process of resuspension is preferentially selective to non-contaminated particles on these atolls to the extent that an aerosol plutonium dilution of 1.8:1 normally occurs.

TABLE 8. Plutonium Aerosol Concentrations on Bikini and Enewetak Atolls Compared (Winds 4-5 m s^{-1}).

Location	Surface Description	Plutonium Aerosol Concentration (aCi m^{-3})	Surface Soil Plutonium Activity (pCi g^{-1})	Estimated Enhancement Factor*
Normal "Background"				
Bikini	Coconut Grove	60	8.01	0.41
Bikini	Stabilized Bare Soil	264	15.3	0.82
Engebi	Vegetated Field	240	24.2	0.45
Unusual Conditions				
Bikini	Field, freshly tilled	6466	15.3	3.10
Engebi	Garden, freshly tilled	7420	24.2	4.41
Engebi	Garden, 1 wk after tilled	3060	24.2	2.55
Bikini	Road with traffic	421	4.10	2.50
Engebi	Downwind of road	1090	35.2	0.56

*Calculated by assuming 34 μg m^{-3} sea spray which has been verified by measurement on Bikini.

During unusual surface conditions, such as immediately after tilling, plutonium aerosol activity (normalized by means of the enhancement factor) also agree well. The corresponding enhancement factors were 4.41 on Engebi Island and 3.10 on Bikini Island.

Plutonium resuspension fluxes due to continuous wind erosion and resuspension were estimated for Bikini by a meteorological flux-gradient equation to be a minimum of 0.49 pCi m^{-2} $year^{-1}$ in the coconut grove and 6.3 pCi m^{-2} $year^{-1}$ over a bare field stabilized by light rain. Since fields do not remain unvegetated for more than a few months, the coconut grove resuspension flux is probably representative of the Island as a whole, even though the wind speeds are one-fourth as high in the coconut grove canopy as in the open.

Particle size distributions measured by both optical and cascade impactor methods show that over the rain-stabilized bare field, the total aerosol size distribution is log-normal with median aerodynamic diameter of 2.44 μm and geometric standard deviation of 3.0, but there is no significant size difference between aerosol plutonium activity and aerosol mass concentration. During the unusual condition of tilling, the size distribution significantly shifts from a median aerodynamic diameter of 2.44 μm to about 2.0 μm with a concurrent increase in plutonium enhancement factor from less than one to 3.1 on Bikini (4.4 on Engebi) and an increase in the pulmonary respirable-fraction from 0.19 to 0.24. In the case of a soil disturbed by tilling on Bikini, the plutonium concentration increased by a factor of 19.1 due to a 3.23 x increase in enhancement factor and a 5.91 x increase in dust aerosol concentrations.

Vehicular traffic produced dust pulses of nominal 10 s duration in a 4.5 m s^{-1} wind, which were log-normally distributed having time-averaged concentrations above background of 28 μg m^{-3} less than 50% of the time and 211 μg m^{-3} less than 5% of the time. (Peak concentrations were a factor of 3.6 higher.) The plutonium enhancement factors for vehicular traffic was 2.5. Foot and bicycle traffic produced dust pulses about one-fourth as large as vehicular traffic.

Personal dosimetry showed that under various exposure conditions, workers inhaled different fractions of inorganic dust and salt, while the organic fraction remained constant at about 10%. Consequently, a personal dosimeter enhancement factor was defined to express the effect a worker has by stirring up dust in his own immediate environment. As a result, pulmonary deposition of plutonium could be calculated for various exposure conditions as determined by inhalation rate, aerosol dust concentration, plutonium activity in soil, plutonium enhancement factor, personal dosimeter enhancement factor, and the pulmonary respirable-fraction. Under the worst case (during tilling), pulmonary deposition was 1476 aCi h^{-1}, and in the best case (light work in a coconut grove), it was 12 aCi h^{-1}. Intermediate cases were for heavy outdoor work (139 aCi h^{-1}) and for light work in and around houses (78 aCi h^{-1}). Walking along a road produced an exposure of 1.58 aCi h^{-1} above background for one vehicular pass per hour. But even with a worst case exposure scenario for 30 years, the lung dose and bone dose due to inhalation on Bikini are well below present Federal Radiation Guidelines.

In conclusion, this study has been the most comprehensive to date, in providing the key parameters for inhalation dose assessment of exposure to plutonium

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contaminated aerosols. Preliminary dose assessments have been verified now by aerosol measurement methods and at different locations. There remain several unexplained and untested results. It is not yet clear why the aerosol dust concentration is apparently uniform for different surface cover and wind conditions (e.g., coconut grove versus bare field). It is also not known why the plutonium enhancement factor is less than unity in the normal case, while at the same time, the aerosol plutonium activity and the aerosol mass size distributions are not significantly different. Long-term monitoring on these remote atolls is not yet very practical, but, some attempts will be made in the near future to monitor meteorological and aerosol levels by the use of satellite telemetry from remote data acquisition systems.

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